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14. ABSTRACT This grant led to considerable successes in our work on LAO/STO interfaces. Our efforts have led to two important discoveries, and there are clear paths to follow for subsequent research. Along with our collaborators, we have constructed LAO/STO structures that, for the first time, clearly exhibit depletion upon adjusting a top-gate voltage. Moreover, we discovered these structures can display a very large					
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a. REPORT UU	b. ABSTRACT UU	c. THIS PAGE UU			19b. TELEPHONE NUMBER 617-253-5585

Report Title

Extremely High Resolution Spectroscopy of Oxide Electronic Systems

ABSTRACT

This grant led to considerable successes in our work on LAO/STO interfaces. Our efforts have led to two important discoveries, and there are clear paths to follow for subsequent research.

Along with our collaborators, we have constructed LAO/STO structures that, for the first time, clearly exhibit depletion upon adjusting a top-gate voltage. Moreover, we discovered these structures can display a very large capacitance enhancement that appears to arise from “negative compressibility” of the electronic system.

Using torque magnetometry, we discovered another surprising feature of the LAO/STO system. We found that the samples exhibited a strong super-paramagnetic signal, originating from the interface, that coexisted with superconductivity. The magnetism was about 0.3-0.4 Bohr Magnetons per unit cell – extremely strong, and it may be indicative of an unusual order parameter in the superconductor.

Each of these results has led to interesting questions (detailed below) that we would like to answer. Theorists have written papers in response to our work on magnetism of the LAO/STO interface, and there are competing models that we would like to test in future work.

Enter List of papers submitted or published that acknowledge ARO support from the start of the project to the date of this printing. List the papers, including journal references, in the following categories:

(a) Papers published in peer-reviewed journals (N/A for none)

<u>Received</u>	<u>Paper</u>
01/29/2013	1.00 C. Richter, Lu Li, S. Paetel, T. Kopp, J. Mannhart, R.C. Ashoori. Very Large Capacitance Enhancement in a Two-Dimensional Electron System, Science, (05 2011): 825. doi:
01/29/2013	2.00 Lu Li, C. Richter, J. Mannhart, R.C. Ashoori. Coexistence of magnetic order and two-dimensional superconductivity at LaAlO ₃ /SrTiO ₃ interfaces, Nature Physics, (10 2011): 762. doi:
TOTAL:	2

Number of Papers published in peer-reviewed journals:

(b) Papers published in non-peer-reviewed journals (N/A for none)

<u>Received</u>	<u>Paper</u>
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TOTAL:

Number of Papers published in non peer-reviewed journals:

(c) Presentations

Number of Presentations: 3.00

Non Peer-Reviewed Conference Proceeding publications (other than abstracts):

<u>Received</u>	<u>Paper</u>
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TOTAL:

Number of Non Peer-Reviewed Conference Proceeding publications (other than abstracts):

Peer-Reviewed Conference Proceeding publications (other than abstracts):

<u>Received</u>	<u>Paper</u>
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TOTAL:

Number of Peer-Reviewed Conference Proceeding publications (other than abstracts):

(d) Manuscripts

<u>Received</u>	<u>Paper</u>
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TOTAL:

Number of Manuscripts:

Books

Received

Paper

TOTAL:

Patents Submitted

Patents Awarded

Awards

Fellow of the American Physical Society (November 2009)

Graduate Students

<u>NAME</u>	<u>PERCENT SUPPORTED</u>	Discipline
Mingda Li	0.15	
FTE Equivalent:	0.15	
Total Number:	1	

Names of Post Doctorates

<u>NAME</u>	<u>PERCENT SUPPORTED</u>	
Lu Li	0.00	
FTE Equivalent:	0.00	
Total Number:	1	

Names of Faculty Supported

<u>NAME</u>	<u>PERCENT SUPPORTED</u>	National Academy Member
Raymond Ashoori	0.08	
FTE Equivalent:	0.08	
Total Number:	1	

Names of Under Graduate students supported

<u>NAME</u>	<u>PERCENT SUPPORTED</u>	Discipline
Scott Johnston	0.00	Physics
Rishi Patel	0.00	Physics
FTE Equivalent:	0.00	
Total Number:	2	

Student Metrics

This section only applies to graduating undergraduates supported by this agreement in this reporting period

The number of undergraduates funded by this agreement who graduated during this period:	1.00
The number of undergraduates funded by this agreement who graduated during this period with a degree in science, mathematics, engineering, or technology fields:.....	1.00
The number of undergraduates funded by your agreement who graduated during this period and will continue to pursue a graduate or Ph.D. degree in science, mathematics, engineering, or technology fields:.....	1.00
Number of graduating undergraduates who achieved a 3.5 GPA to 4.0 (4.0 max scale):	1.00
Number of graduating undergraduates funded by a DoD funded Center of Excellence grant for Education, Research and Engineering:.....	0.00
The number of undergraduates funded by your agreement who graduated during this period and intend to work for the Department of Defense	0.00
The number of undergraduates funded by your agreement who graduated during this period and will receive scholarships or fellowships for further studies in science, mathematics, engineering or technology fields:	0.00

Names of Personnel receiving masters degrees

NAME

Total Number:

Names of personnel receiving PhDs

NAME

Total Number:

Names of other research staff

NAME

PERCENT SUPPORTED

FTE Equivalent:

Total Number:

Sub Contractors (DD882)

Inventions (DD882)

Scientific Progress

Large Capacitance Enhancement in LAO/STO Interfaces

In the simple geometry shown in Figure 1, we have made capacitance measurements with a gold or YBCO metallic disk as a top electrode in a capacitor with the electronic system at the LAO/STO interface as the bottom electrode. We performed these experiments with several different samples with LAO thicknesses of 10 and 12 unit cells. To our surprise, we were able to gate the samples into full depletion using the top gate despite our initial thought that the charge density at the interface would be too large to do this. It may be that the strain from the top gate or the work function between the YBCO top gate and STO acted to diminish the electron density at the interface sufficiently to allow depletion. Whatever the reason, the ability to achieve full depletion allowed us to make measurements of an important thermodynamic quantity (the electronic compressibility) in the low-density regime where electron-electron interaction energies grow to be comparable or larger than the Fermi energy.

In the low-density regime in two-dimensional semiconductor electronic systems, theorists predicted and subsequent measurements demonstrated that the electronic compressibility of the system can actually reverse sign to become negative at low densities. This happens as exchange and correlation energies become larger than quantum single particle energies at low densities. As exchange and correlation are attractive interactions, this diminishes the energy cost for adding an electron to the system. In fact, the energy cost for adding an electron to the system can drop below the classical charging energy (the cost for adding a charge uniformly smeared across a capacitor plate). This phenomenon is known as “negative compressibility”, and it results the measured capacitance between the plates growing larger than the value expected from the geometry of the capacitor and the dielectric constant of the material between the plates. This effect is shown in Figure 2.

In semiconductor samples, the observed capacitance enhancement is one or two percent, and this is only observed in the highest mobility samples. In contrast, in the LAO/STO system, we have observed enhancements of greater than 40% (although other samples show smaller enhancements). Moreover, we believe that the actual enhancement is greater than the observed 40%. As the bias on the top gate is moved closer to depletion (at a top gate voltage of around 0.30 Volts in Figure 2), the sample resistivity diverges (so that we cannot adequately charge to sample due to the large RC time for charging), preventing us from observing the capacitance in this regime. The observation of the large capacitance enhancement is remarkable both for the large magnitude of the effect, the observed enhancement at room temperature (see Publication [1]), and the fact that the enhancement exists in a highly disordered sample. While the origin of the effect may lie in the same exchange enhancement that creates negative compressibility in semiconductors, we have not proven this, and curve-fitting using a model of exchange enhancement fails. The capacitance upturn in Figure 2 is too sudden to be fit by this model.

We are left with the question of just how large the actual increase in the capacitance might be. Answering this question might give us a better idea of the origin of the effect. One possibility is to measure the capacitance at lower frequencies. We have tried this, but the $1/f$ noise from our amplifiers has made this practically impossible. The solution is to diminish the charging time of the sample. For future experiments, our collaborators have grown a new sample for us that is pictured in Figure 3.

The new sample is designed for rapid charging to allow us to examine in detail the possible divergence of the capacitance at low densities. The basic idea is to minimize the distance that charge will need to travel to allow full charging of the capacitor at sufficiently high frequencies so that our measurements are not swamped with $1/f$ noise. Due to the high dielectric constant (18) and the small thickness of the LAO material, the small pads in shown in Figure 3 will still have capacitance greater than 100 picofarads. This will allow for relatively simple capacitance measurements that will not require a low temperature amplifier.

The sample geometry shown in Figure 3 will allow us to build quantum dots in this material. With luck, we should be able to build tiny capacitors and observe charging starting from the first electron in the capacitor. Observing the capacitance divergence as single electrons enter the material, one by one, may give us powerful clues to the origin of the negative compressibility. For instance, we may find (as we have seen in semiconductors – and has still not been explained) that electrons enter the material in bunches rather than the usual pattern of Coulomb Blockade.

As we explained in a publication on this effect[1], understanding this problem could be of significant technological interest. The capacitor has large capacitance at low densities, and this is precisely what would be required to switch transistors using very low voltages. As the dissipation in computer circuits depends on the square of the voltage, understanding and exploitation of this effect (persisting at room-temperature!) could have tremendous practical implications. Finally, we would like to ultimately use our pulsed tunneling techniques (tunneling through the LAO) to directly observe the band structure at the interface and how it changes with electron density.

Coexistence of Magnetism and Superconductivity in LAO/STO interfaces.

Using torque magnetometry, we have discovered a strong paramagnetism (or soft ferromagnetism) that coexists with the superconducting state in LAO/STO. Superconductivity and magnetic order are in general mutually exclusive phenomena with exceptions for exotic cases in three dimensional systems such as $\text{RuSr}_2\text{GdCu}_2\text{O}_8$ (perhaps a triplet pairing state) and in the

heavy fermion systems UGe₂ (thought to be a finite momentum pairing – FFLO state). Before our work and that of 2 other competing groups it was unknown whether such coexistence could occur in a two-dimensional electronic system. Figure 4 shows results that are typical of our samples. It displays a clear coexistence of superconductivity and magnetism. The strength of the magnetism is quite large, around 0.3 Bohr magnetons per unit cell. If the ferromagnetism and superconductive both occur at the interface (as our measurements lead us to believe – see reference [1]), the results make us wonder if the superconductivity must be of some unconventional form in order to coexist with the ferromagnetism.

Michaeli, Potter, and Lee (MPL) have presented a more conventional (although still unusual) picture from MIT. They have developed a model about what may be giving rise to our unusual results (see arXiv:1107.4352v). Essentially, they propose a model of the interface in which the mobile electrons do not arise from the polar catastrophe model (reference). As the polar catastrophe model predicts a charge density at the LAO/STO interface consisting of $\frac{1}{2}$ of an electron per unit cell, their model describes these electrons as being in an ordered state and localized by strong interactions (a Mott Insulator). The electrons responsible for conduction are thought to arise from either oxygen vacancies or from deviations from $\frac{1}{2}$ electron per unit cell in the polar catastrophe model. These extra electrons do not fit in the same the same layer of dxy orbitals as those right at the interface. Instead, electrostatics places them in dxy orbitals in adjacent layers (see Figure 5). With enough excess electrons some of them will enter dxz and dyz orbitals. In the MPL model, electrons in these bands have a strong exchange coupling with electrons in the interfacial dxy orbitals. Because there is a low density of electrons in the dxz and dyz orbitals, the effective Fermi wavelength is long, allowing for long range RKKY coupling between spins in the interfacial layer that produces ferromagnetism in the interfacial layer. MPL describe the superconductivity as occurring in the dxz and dyz orbitals. Due to a large Rashba term in the spin-orbit coupling the superconductivity can exist in an FFLO state (finite momentum pairing).

Tests of MPL the model

The MPL model results in a simple prediction that we should be able to test. If indeed the conduction electrons are in the dxz and dyz orbitals this might explain why the measured charge densities in all experiments are about at least an order of magnitude smaller than expected in the polar catastrophe model. Depleting the electrons in the dxz and dyz orbitals by means of gating should make magnetism disappear. In future work, we will build samples for our cantilevers that will allow gating, and our plan is to monitor the magnetism as we vary the electron density. As it is easy to fabricate samples for back-gating, we will try these first. However, back-gating does not appear to allow for full depletion of conduction electrons. Moreover, we may face some difficulties in bringing the large voltages for back gating through a fine wire attached to a sample mounted on a delicate metallic cantilever. For these reasons, we will also create samples for top gating. From our previous experience, we know that we can fully deplete conduction electrons with a top gate. As large top gates tend to leak, these samples will need to be small. This will require enhancement of the sensitivity of the current set-up. For this, we plan to use a low temperature amplifier for capacitive readout of our cantilevers.

Technology Transfer

Final Technical Report

Extremely High Resolution Spectroscopy of Oxide Electronic Systems

Summary:

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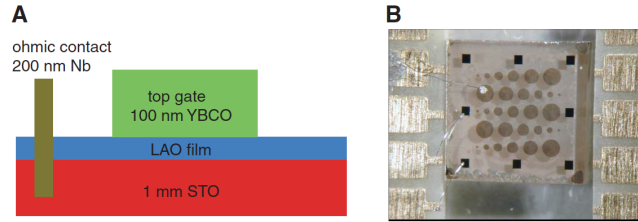
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Details of work:

Large Capacitance Enhancement in LAO/STO Interfaces

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Figure 1, A: Basic structure of samples for capacitance / compressibility experiment. B: A photograph of one of the samples used for the experiment. The dark squares around the edges of the sample are the niobium contacts. The gold circles are the top gates.



In the low-density regime in two-dimensional semiconductor electronic systems, theorists predicted and subsequent measurements demonstrated that the electronic compressibility of the system can actually reverse sign to become negative at low densities. This happens as exchange and correlation energies become larger than quantum single particle energies at low densities. As exchange and correlation are attractive interactions, this diminishes the energy cost for adding an electron to the system. In fact, the energy cost for adding an electron to the system can drop below the classical charging energy (the cost for adding a charge uniformly smeared across a capacitor plate). This phenomenon is known as “negative compressibility”, and it results the measured capacitance between the plates growing larger than the value expected from the geometry of the capacitor and the dielectric constant of the material between the plates. This effect is shown in Figure 2.

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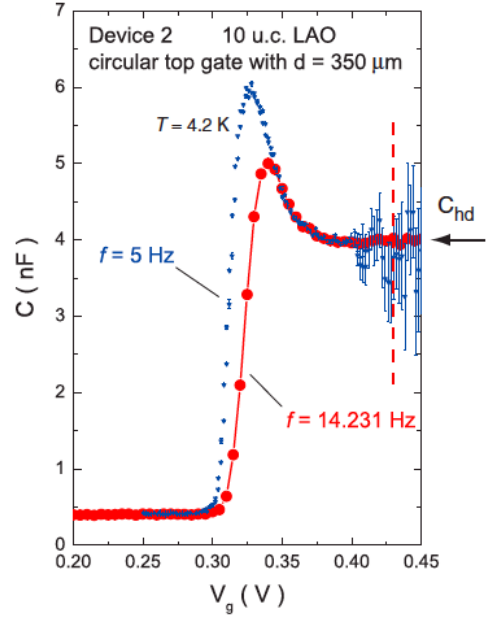


Figure 2. Capacitance signals from a sample of the type shown in Figure 1. Here, full depletion occurs at about 0.3 Volts. The sample becomes very resistive near full depletion, leading to a frequency dependence in the capacitance signal. Measurements at yet lower frequencies may show a considerably larger enhancement of the capacitance beyond the 40% enhancement of the capacitance seen in the blue curve.

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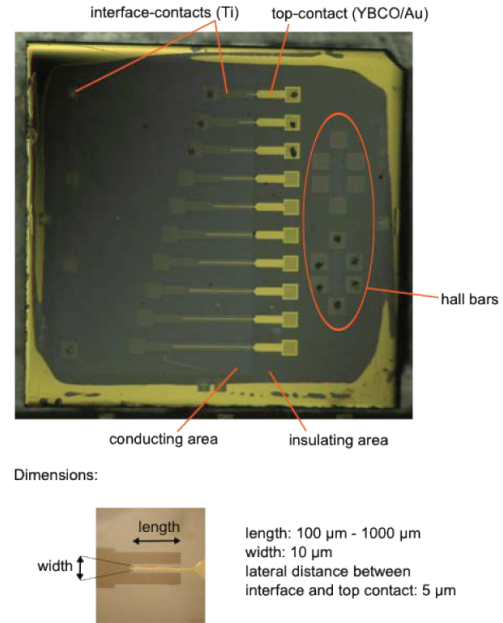


Figure 3: View of the new sample for capacitance and compressibility measurements. We have designed the long and narrow capacitors are to charge even under conditions of very large in-plane resistance. This sample should allow us to explore the capacitance divergence at low densities in much greater detail.

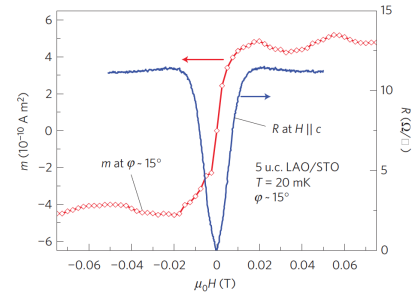


Figure 4: Magnetization (red) and resistivity (blue) of an LAO/STO sample. Notice the coexistence of magnetism with superconductivity.

adjacent layers (see Figure 5). With enough excess electrons some of them will enter d_{xz} and d_{yz} orbitals. In the MPL model, electrons in these bands have a strong exchange coupling with electrons in the interfacial d_{xy} orbitals. Because there is a low density of electrons in the d_{xz} and d_{yz} orbitals, the effective Fermi wavelength is long, allowing for long range RKKY coupling between spins in the interfacial layer that produces ferromagnetism in the interfacial layer. MPL describe the superconductivity as occurring in the d_{xz} and d_{yz} orbitals. Due to a large Rashba term in the spin-orbit coupling the superconductivity can exist in an FFLO state (finite momentum pairing).

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Publications:

- [1] Lu, Li, C. Richter, S. Paetel, T. Kopp, J. Mannhart, R.C. Ashoori, “Very Large Capacitance Enhancement in a Two-Dimensional Electron System” *Science* **332**, pp. 825-828 (2011)
- [2] Lu, Li, C. Richter, J. Mannhart, R.C. Ashoori, “Coexistence of magnetic order and two-dimensional superconductivity at LaAlO₃/SrTiO₃ interfaces” *Nature Physics* **Vol. 7** pp. 762-766 (2011)

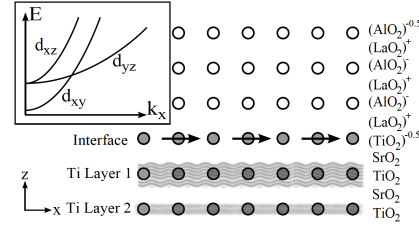


Figure 5. Picture (figure 1) from the MPL paper.